WHERE HIGH RESOLUTION SPECTROSCOPY MEETS DYNAMICS: FROM MOLECULAR CRASHES TO SPLASHES

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High resolution laser spectroscopy offers a sensitive and universal method for detecting molecules at the quantum state level, which can prove remarkably powerful in facilitating detailed studies of fundamental chemical physics phenomena. Time permitting, this talk will present examples from our group in several complementary areas which exploit high resolution laser methods to probe collision dynamics at the state-to-state level. i) Reactive scattering in crossed molecular beams will be discussed, based on intense pulsed discharge radical atom sources and probing rovibrational levels of the nascent products via cw direct IR laser absorption. Studies of elementary F atom abstraction will be used to illustrate simple models for atom + diatom, triatom and polyatom reaction systems, the last of which highlight surprisingly strong quantum state resolved correlations between translational energy recoil and rovibrational excitation of the nascent products. ii) A novel spectroscopic method for studying molecular dynamics at the gas-liquid interface will also be discussed. This approach is based on colliding a supersonic beam of jet cooled molecules onto a continuously renewable liquid surface in vacuum, taking advantage of high resolution IR laser Dopplerimetry to probe the nascent rotational, vibrational and translational distributions of scattered species. This provides a wealth of additional dynamical information, and is quite complementary to the translational kinetic energy studies by Nathanson and coworkers. The method will be illustrated by recent results from our group on jet cooled CO2 ”splashing” from liquids such as perfluorpolyether (PFPE) and squalane, as a function of center of mass collision energy. The results indicate non-equilibrium superthermal distributions in the scattered states, and provide independent support for dual trapping desorption (TD) and impulsive scattering (IS) channels in the gas-liquid collision event.